Intermittent atomic fluorescence

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In a single-atom double-resonance experiment involving a strong and a weak transition, quantum jumps on the weak transition cause the resonance fluorescence of the strong transition to turn on and off randomly in time when observed over intervals long compared to the lifetime of the strong transition but short compared to that of the weak transition. Here we present a theory of this intermittent atomic fluorescence for the case of coherent excitation.

I. INTRODUCTION

The observation of photon antibunching in single-atom resonance fluorescence has provided confirmation of the discrete nature of quantum transitions in an individual quantum system.[1-3] Similar observations of the tunneling of individual electrons in metal-oxide-semiconductor junctions[4] and of spin flips of individual electrons in a Penning trap[5] provide further confirmation of such discrete transitions. Recently there has been considerable interest in a scheme suggested by Dehmelt that could provide a remarkably direct means of observing the quantum nature of optical transitions in an atom.[6-8]

Consider an atom having strong and weak transitions with a common lower level, as depicted in Fig. 1. With the strong transition illuminated by an external light source and no radiation acting on the weak transition, the atom fluoresces strongly due to spontaneous emission from level 1. The atom can repeatedly undergo transitions from level 0 to level 1 and back to level 0, giving rise to, say, $A_1 \sim 10^8$ fluorescent photons per second. This fluorescence, of course, exhibits photon antibunching. If we now apply radiation to the weak transition, the electron is occasionally promoted to level 2 and the strong fluorescence is interrupted because the electron is unavailable for transitions between levels 0 and 1. It is said that the electron is "shelved" in level 2.[6] The fluorescence can resume when the electron returns to level 0 either by spontaneous or stimulated emission. For the sake of argument, a residence time A_2^{-1} in level 2 on the order of 1 sec is not unreasonable, making for a tremendous disparity in time scales ($A_1^{-1} \ll A_2^{-1}$). Because of this disparity and since we are principally interested in the interruptions of the strong fluorescence caused by the weak transition, we will adopt a point of view in which time is coarse-grained over intervals long compared with A_1^{-1} but short compared with A_2^{-1}. This procedure naturally removes certain nonclassical features such as antibunching of the light from the strong transition and effectively converts the temporal variations of the strong fluorescence into a classical stochastic process. We will see that such a point of view is fully justified from the density-matrix equations of motion. Because the transitions $0 \leftrightarrow 2$ occur randomly in time, the atomic fluorescence resembles a classical random telegraph signal.

The effect just described is currently of interest because (i) it should be accessible to experiment in the near future through single-ion trapping techniques,[9-11] (ii) it provides a means by which the quantum jumps of a single electron in a single atom or ion can be directly monitored,[7] (iii) it supplies a technique for the detection of very weak atomic lines,[12] and (iv) it poses a number of challenging theoretical problems, especially as regards quantum-measurement theory.

In a recent publication we presented a simple theory of the intermittent fluorescent signal for the case of incoherent excitation,[7] i.e., the case in which the spectral energy density of the exciting radiation is a slowly varying function of frequency across each atomic line. This special case was chosen for mathematical convenience. For incoherent excitation, the populations of atomic states are governed by the Einstein rate equations, which are simple. However, this case does not apply to experiments in which coherent laser excitation is used. In this case, one expects a number of coherent effects such as Rabi oscillations and Autler-Townes splitting of the weak atomic line. It is the

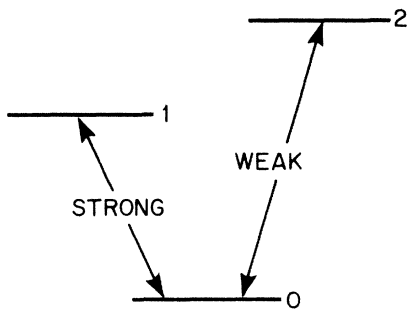

FIG. 1. Energy-level scheme for intermittent fluorescence.

purpose of this paper to present a theory of intermittent atomic fluorescence for the case of coherent excitation, a theory that is needed for the planning and interpretation of experiment.

It turns out that the results for coherent excitation are surprisingly similar to those for incoherent excitation. The simplicity of the theory for incoherent excitation derives from the fact that the excitation of the weak transition is a rate process. That is, there exist probabilities per unit time R_+ and R_- that the electron makes upward or downward jumps on this transition. What we show here is that for fairly general conditions of coherent excitation of the strong transition, the excitation of the weak transition is again describable as a rate process. Hence much of the earlier theory applies directly to the coherent case. The only substantive change is that the transition rates R_+ and R_- are expressed in terms of coherent parameters, such as Rabi frequencies, instead of spectral energy densities and Einstein B coefficients.

II. COHERENT ATOMIC DYNAMICS

To show that excitation of the weak transition is a rate process, it is sufficient to show that this excitation is accurately described by rate equations rather than Bloch equations or density-matrix equations. The quantum-regression theorem then ensures that higher-order atomic correlation functions will likewise be describable as a rate process. To this end we start with the full density-matrix equations for coherent excitation of the three-level atom. In the electric dipole[13] approximation these equations are[14]

$$\dot{\rho}_{11}=-A_1\rho_{11}+i(\rho_{01}-\rho_{10})\mu_1E(t)/\hbar\,, \tag{1a}$$

$$\dot{\rho}_{22}=-A_2\rho_{22}+i(\rho_{02}-\rho_{20})\mu_2E(t)/\hbar\,, \tag{1b}$$

$$\dot{\rho}_{00}=A_1\rho_{11}+A_2\rho_{22}-i(\rho_{01}-\rho_{10})\mu_1E(t)/\hbar -i(\rho_{02}-\rho_{20})\mu_2E(t)/\hbar\,, \tag{1c}$$

$$\dot{\rho}_{01}=(i\omega_{10}-\tfrac{1}{2}A_1)\rho_{01}+i(\rho_{11}-\rho_{00})\mu_1E(t)/\hbar +i\mu_2\rho_{21}E(t)/\hbar\,, \tag{1d}$$

$$\dot{\rho}_{02}=(i\omega_{20}-\tfrac{1}{2}A_2)\rho_{02}+i(\rho_{22}-\rho_{00})\mu_2E(t)/\hbar +i\mu_1\rho_{12}E(t)/\hbar\,, \tag{1e}$$

$$\dot{\rho}_{12}=(i\omega_{21}-\tfrac{1}{2}A_1-\tfrac{1}{2}A_2)\rho_{12} +i(\mu_1\rho_{02}-\mu_2\rho_{10})E(t)/\hbar\,. \tag{1f}$$

Here A_1 and A_2 are the Einstein spontaneous-emission rates from levels 1 and 2 to level 0; ω_{10}, ω_{20}, and ω_{21} are the three transition frequencies; μ_1 and μ_2 are the dipole transition moments for transitions $0\leftrightarrow1$ and $0\leftrightarrow2$, respectively, and

$$E(t)=\varepsilon_1\cos(\omega_1t)+\varepsilon_2\cos(\omega_2t)\,, \tag{2}$$

is the applied electric field with components at frequencies ω_1 and ω_2, which are near to resonance with the transition frequencies ω_{10} and ω_{20}, respectively. The diagonal elements of the density matrix ρ_{00}, ρ_{11}, and ρ_{22} are the probabilities P_0, P_1, and P_2 that levels 0, 1, and 2 are occupied. For the off-diagonal elements or "coherences" it is convenient to make the substitutions

$$\rho_{01}=\sigma_{01}e^{i\omega_1t}\,, \tag{3a}$$

$$\rho_{02}=\sigma_{02}e^{i\omega_2t}\,, \tag{3b}$$

$$\rho_{12}=\sigma_{12}e^{i(\omega_2-\omega_1)t}\,, \tag{3c}$$

which allow us to work with the slowly changing variables σ_{01}, σ_{02}, and σ_{12}. On substituting (3) into (1) and using (2) we obtain equations for P_0, P_1, P_2, σ_{01}, σ_{02}, and σ_{12} that contain slowly varying terms and rapidly oscillating terms. The latter are negligible because they average to zero over very short time intervals of order $1/\omega_1$, $1/\omega_2$, or $1/(\omega_2-\omega_1)$. In the rotating-wave approximation, the rapidly oscillating terms are discarded, and Eqs. (1) reduce to

$$\dot{P}_1=-A_1P_1+\tfrac{1}{2}i\Omega_1(\sigma_{01}-\sigma_{10})\,, \tag{4a}$$

$$\dot{P}_2=-A_2P_2+\tfrac{1}{2}i\Omega_2(\sigma_{02}-\sigma_{20})\,, \tag{4b}$$

$$\dot{P}_0=A_1P_1+A_2P_2-\tfrac{1}{2}i\Omega_1(\sigma_{01}-\sigma_{10}) -\tfrac{1}{2}i\Omega_2(\sigma_{02}-\sigma_{20})\,, \tag{4c}$$

$$\dot{\sigma}_{01}=-(i\Delta_1+\tfrac{1}{2}A_1)\sigma_{01}+\tfrac{1}{2}i\Omega_1(P_1-P_0) +\tfrac{1}{2}i\Omega_2\sigma_{21}\,, \tag{4d}$$

$$\dot{\sigma}_{02}=-(i\Delta_2+\tfrac{1}{2}A_2)\sigma_{02}+\tfrac{1}{2}i\Omega_2(P_2-P_0) +\tfrac{1}{2}i\Omega_1\sigma_{12}\,, \tag{4e}$$

$$\dot{\sigma}_{12}=-[i(\Delta_2-\Delta_1)+\tfrac{1}{2}(A_1+A_2)]\sigma_{12} +\tfrac{1}{2}i\Omega_1\sigma_{02}-\tfrac{1}{2}i\Omega_2\sigma_{10}\,, \tag{4f}$$

where $\Delta_1=\omega_1-\omega_{10}$ and $\Delta_2=\omega_2-\omega_{20}$ are the detunings on the strong and weak transitions, respectively, and

$$\Omega_1=\mu_1\varepsilon_1/\hbar\,, \tag{5a}$$

$$\Omega_2=\mu_2\varepsilon_2/\hbar\,, \tag{5b}$$

are the on-resonance Rabi frequencies for these two transitions.

A transformation from density-matrix components to real Bloch variables is convenient for the subsequent analysis. The three-level Bloch variables u_1,v_1, u_2,v_2, and u_3,v_3 are twice the real and imaginary parts of the coherences σ_{01}, σ_{02}, and σ_{12},

$$\sigma_{01}=\tfrac{1}{2}(u_1+iv_1)\,, \tag{6a}$$

$$\sigma_{02}=\tfrac{1}{2}(u_2+iv_2)\,, \tag{6b}$$

$$\sigma_{12}=\tfrac{1}{2}(u_3+iv_3)\,. \tag{6c}$$

In addition, we shall work with the inversion on the strong transition

$$w_1=P_1-P_0\,, \tag{7}$$

and the probabilities

$$\mathscr{P}_+=P_2\,, \tag{8a}$$

$$\mathscr{P}_-=P_0+P_1\,. \tag{8b}$$

$\mathscr{P}_+$ is the probability that the weak transition is excited and $\mathscr{P}_-$ the probability that it is not excited. In terms of these new variables Eqs. (4) read

$$\dot{u}_1=\Delta_1 v_1-\tfrac{1}{2}A_1 u_1+\tfrac{1}{2}\Omega_2 v_3 , \tag{9a}$$

$$\dot{v}_1=-\Delta_1 u_1-\tfrac{1}{2}A_1 v_1+\Omega_1\omega_1+\tfrac{1}{2}\Omega_2 u_3 , \tag{9b}$$

$$\dot{\omega}_1=-\Omega_1 v_1-A_1(\mathscr{P}_-+\omega_1)-A_2\mathscr{P}_+-\tfrac{1}{2}\Omega_2 v_2 ; \tag{9c}$$

$$\dot{u}_3=(\Delta_2-\Delta_1)v_3-\tfrac{1}{2}(A_1+A_2)u_3-\tfrac{1}{2}\Omega_1 v_2-\tfrac{1}{2}\Omega_2 v_1 , \tag{10a}$$

$$\dot{v}_3=-(\Delta_2-\Delta_1)u_3-\tfrac{1}{2}(A_1+A_2)v_3+\tfrac{1}{2}\Omega_1 u_2-\tfrac{1}{2}\Omega_2 u_1 ; \tag{10b}$$

$$\dot{u}_2=\Delta_2 u_2-\tfrac{1}{2}A_2 u_2-\tfrac{1}{2}\Omega_1 v_3 , \tag{11a}$$

$$\dot{v}_2=-\Delta_2 u_2-\tfrac{1}{2}A_2 v_2+\tfrac{1}{2}\Omega_1 u_3+\Omega_2(\mathscr{P}_+-\tfrac{1}{2}\mathscr{P}_-+\tfrac{1}{2}w_1) ; \tag{11b}$$

$$\dot{\mathscr{P}}_+=-A_2\mathscr{P}_+-\tfrac{1}{2}\Omega_2 v_2 , \tag{12a}$$

$$\dot{\mathscr{P}}_-=A_2\mathscr{P}_-+\tfrac{1}{2}\Omega_2 v_2 . \tag{12b}$$

When we say that the transition $0\leftrightarrow1$ is strong and $0\leftrightarrow2$ is weak, we mean that the spontaneous and induced transition rates on $0\leftrightarrow1$ are much larger than the spontaneous or induced rates on $0\leftrightarrow2$:

$$A_1,\Omega_1\gg A_2,\Omega_2 . \tag{13}$$

This being the case, we see that the last terms in (9a) and (9b) and the last two terms in (9c) are negligible compared to other terms in these equations (the Bloch variables and the probabilities $\mathscr{P}_\pm$ are nominally of order unity or less.) Hence Eqs. (9) reduce to

$$\dot{u}_1=\Delta_1 v_1-\tfrac{1}{2}A_1 u_1 , \tag{14a}$$

$$\dot{v}_1=-\Delta_1 u_1+\Omega_1 w_1-\tfrac{1}{2}A_1 v_1 , \tag{14b}$$

$$\dot{w}_1=-\Omega_1 v_1-A_1(\mathscr{P}_-+w_1) . \tag{14c}$$

These are just the usual Bloch equations for a two-level system (levels 0 and 1), except that the probability to be in one or the other of these levels is $\mathscr{P}_-$ instead of unity. Because of the large relaxation rate A_1 (say 10^8 sec^{-1}) in (14), the Bloch variables u_1, v_1, and w_1 relax to their steady-state values in a very short time ($\sim10^{-8}$sec), and, if $\mathscr{P}_-$ varies slowly (say $\dot{\mathscr{P}}_-\sim1$ sec^{-1}), the Bloch variables adiabatically follow this variation to an excellent approximation. That is, the Bloch variables are at each instant given by the steady-state solution of (14)—the solution obtained by solving (14) with $\dot{u}_1=\dot{v}_1=\dot{w}_1=0$. The simplify the algebra we shall assume that the strong transition is driven on resonance ($\Delta_1=0$). In this case, the steady-state solution of Eqs. (14) is

$$u_1=0 , \tag{15a}$$

$$v_1=-2A_1\Omega_1\mathscr{P}_-/D_1 , \tag{15b}$$

$$w_1=-A_1^2\mathscr{P}_-/D_1 , \tag{15c}$$

where

$$D_1=A_1^2+2\Omega_1^2 . \tag{16}$$

Note that, for very short times of order A_1^{-1}, the adiabatic solution (15) may differ from the exact solution to Eqs. (14) when the initial conditions on u_1, v_1, and w_1 differ from the adiabatic values (15). Here and in the following we ignore such rapid transients which are invisible on the course-grained time scale of interest.

Similarly, the Bloch variables u_3 and v_3 of Eqs. (10) adiabatically follow the inhomogeneous terms in these equations because the relaxation rate $(A_1+A_2)/2$ is large. In this rate, A_2 is negligible compared to A_1 and may be discarded. Then the steady-state solution of Eqs. (10), with u_1 and v_1 taken from (15) and $\Delta_1=0$, is

$$u_3=(2\Delta_2\Omega_1 u_2-A_1\Omega_1 v_2)/D_2+2A_1^2\Omega_1\Omega_2\mathscr{P}_-/D_1D_2 , \tag{17a}$$

$$v_3=(A_1\Omega_1 u_2+2\Delta_2\Omega_1 v_2)/D_2-4A_1\Delta_2\Omega_1\Omega_2\mathscr{P}_-/D_1D_2 , \tag{17b}$$

where

$$D_2=4\Delta_2^2+A_1^2 . \tag{18}$$

Use of (15c) and (17) in Eqs. (11) yields the following equations for the Bloch variables of the weak transition:

$$\dot{u}_2=\Delta v_2-\tfrac{1}{2}\gamma u_2+2\Delta_2 A_1\Omega_1^2\Omega_2\mathscr{P}_-/D_1D_2 , \tag{19a}$$

$$\dot{v}_2=-\Delta u_2-\tfrac{1}{2}\gamma v_2+\Omega_2\left[\mathscr{P}_+-\tfrac{1}{2}\left[1+\frac{A_1^2}{D_1}-\frac{2A_1^2\Omega_1^2}{D_1D_2}\right]\mathscr{P}_-\right], \tag{19b}$$

where

$$\Delta=\Delta_2\left[\frac{4\Delta_2^2+A_1^2-\Omega_1^2}{4\Delta_2^2+A_1^2}\right] \tag{20a}$$

is the *effective* detuning for this transition and

$$\gamma=A_2+\frac{A_1\Omega_1^2}{4\Delta_2^2+A_1^2} \tag{20b}$$

is the transverse relaxation rate.

The really important point here is that the radiation applied to the strong transition greatly increases the transverse relaxation rate of the weak transition. This damping is accomplished via Raman-type processes that couple the dipoles σ_{01} and σ_{02} via σ_{12}. In this way the $0\leftrightarrow1$ transition acts as a reservior that damps the coherence of the $0\leftrightarrow2$ transition. When the strong transition is saturated ($\Omega_1\gg A_1$), the second term on the right in (20b) is of order A_1 or larger, except when the detuning Δ_2 is very large ($|\Delta_2|\gg\Omega_1$). This very large relaxation rate causes the Bloch variables u_2 and v_2 to adiabatically follow the inhomogeneous terms in (19), and we have from the steady-state solution

$$v_2=\frac{2\Omega_2\gamma}{4\Delta^2+\gamma^2}(\mathscr{P}_+-\delta\mathscr{P}_-) , \tag{21}$$

where

$$\delta=\tfrac{1}{2}\left[1+\frac{A_1^2}{D_1}-\frac{2A_1^2\Omega_1^2}{D_1D_2}\right]+\frac{4\Delta_2\Delta A_1\Omega_1^2}{\gamma D_1D_2}\ . \tag{22}$$

On substituting this into Eqs. (12) we find that the probabilities $\mathscr{P}_+$ and $\mathscr{P}_-$ obey the rate equations.

$$\dot{\mathscr{P}}_+=-R_-\mathscr{P}_++R_+\mathscr{P}_-\ , \tag{23a}$$

$$\dot{\mathscr{P}}_-=R_-\mathscr{P}_+-R_+\mathscr{P}_-\ , \tag{23b}$$

where the upward and downward transition rates are

$$R_+=\frac{\gamma\Omega_2^2\delta}{4\Delta^2+\gamma^2} \tag{24a}$$

and

$$R_-=A_2+\frac{\gamma\Omega_2^2}{4\Delta^2+\gamma^2}\ , \tag{24b}$$

respectively. This completes the proof that the excitation of the weak transition is a rate process when observed over coarse-grained intervals of time.

To better understand the meaning of these results, we first consider the case in which the strong transition is strongly saturated ($\Omega_1\gg A_1$) and the detuning Δ_2 is approximately $\pm\frac{1}{2}\Omega_2$. Here we anticipate the Autler-Townes splitting of the weak atomic line and assume that the field acting on the weak transition is nearly resonant with one component of the Autler-Townes doublet. In this case

$$\Delta=\Delta_2\left[\frac{4\Delta_2^2-\Omega_1^2}{4\Delta_2^2+A_1^2}\right]\ , \tag{25a}$$

$$\gamma=\frac{A_1\Omega_1^2}{4\Delta_2^2+A_1^2}\ , \tag{25b}$$

and $\delta=\frac{1}{2}$ [in (25b) we have discarded the negligible contribution A_2 to γ]. The upward and downward transition rates are

$$R_+=\frac{2A_1\Omega_1^2\Omega_2^2\Delta_2^2}{4\Delta_2^2(4\Delta_2^2-\Omega_1^2)^2+A_1^2\Omega_1^4}\ , \tag{26a}$$

$$R_-=A_2+2R_+\ . \tag{26b}$$

As a function of Δ_2, R_+ has two sharp peaks at $\Delta_2=\pm\frac{1}{2}\Omega_1$, which are well represented by a pair of Lorentzian functions each of width $A_1/2$,

$$R_+\approx\frac{A_1\Omega_2^2}{8}\left[\frac{1}{(\Delta_2-\frac{1}{2}\Omega_1)^2+(A_1/4)^2}+\frac{1}{(\Delta_2+\frac{1}{2}\Omega_1)^2+(A_1/4)^2}\right]\ . \tag{27}$$

This is the Autler-Townes doublet, here appearing in the upward transition rate.[14] Equation (27) is accurate in the neighborhood of the peaks. However, away from the peaks, where R_+ is small, Eq. (27) is not accurate. For example, at $\Delta_2=0$, Eq. (24a) indicates that $R_+=\Omega_2^2A_1^3/2\Omega_1^4$, while the approximation (27) gives a different result.

Next consider the steady-state solution of the rate equations (23) in this same limit. The probability that the weak transition is excited is

$$\mathscr{P}_+=\frac{R_+}{R_++R_-}=\frac{R_+}{A_2+3R_+}\ , \tag{28}$$

or, in general, $\mathscr{P}_+=R_+/[A_2+R_+(1+1/\delta)]$. This is also the fraction of the time that the fluorescence is off, since the fluorescence is off whenever the atomic electron is in level 2. Suppose we tune to one component of the doublet ($\Delta_2=\pm\frac{1}{2}\Omega_1$). Then $R_+=\Omega_2^2/2A_1$ and $\mathscr{P}_+=\Omega_2^2/(2A_1A_2+3\Omega_2^2)$. For a strong field on the weak transition ($\Omega_2^2\gg A_1A_2$), $\mathscr{P}_+=\frac{1}{3}$ and the fluorescence is off one-third of the time. Note that, in the case under consideration, a Rabi frequency Ω_2 of the order of the geometric mean of A_1 and A_2 is required to make $\mathscr{P}_+$ of order unity.

It is possible to obtain a larger probability for level 2 with a smaller Rabi frequency Ω_2 by decreasing the field strength acting on transition $0\leftrightarrow1$. This may be of importance in experiment, since a full test of the present theory requires a substantial probability $\mathscr{P}_+$, and a large Rabi frequency Ω_2 may be difficult to achieve owing to the small transition dipole μ_2.

To show that the weak transition is more easily excited when the strong one is weakly driven, we consider the case where $\Omega_1<A_1$ and Δ_2 is small in magnitude compared to A_1. The latter condition is sufficient for our purpose because, for $\Omega_1\ll A_1$, there is no splitting of the weak atomic line, and the broadening is much less than A_1. Under these conditions, Eqs. (22) and (24) give $\delta=1$ and

$$R_+=\frac{\Omega_1^2\Omega_2^2}{A_1(4\Delta_2^2+\Omega_1^4/A_1^2)}\ , \tag{29a}$$

$$R_-=A_2+R_+\ . \tag{29b}$$

Observe that the linewidth for upward transitions is Ω_1^2/A_1, which is indeed small compared to A_1 when $\Omega_1\ll A_1$. But Ω_1 must not be made too small. In order for the above analysis to remain valid, the inequality (13) must hold, and γ must still be large ($\gamma\gg A_2,\Omega_2$) so that the adiabatic following approximation is valied in Eqs. (19). These conditions are met when $\Omega_1^2\gg A_1A_2$, $A_1\Omega_2$. On resonance ($\Delta_2=0$), the steady-state level-2 probability is now $\mathscr{P}_+=1/(2+A_2\Omega_1^2/A_1\Omega_2^2)$. This is substantial ($\mathscr{P}_+=\frac{1}{3}$) at $\Omega_2=\Omega_1(A_2/A_1)^{1/2}$, a value much smaller than for a saturated strong transition.

III. STATISTICS OF FLUORESCENCE

Given that the excitation of the weak transition is a rate process, the statistical analysis of the fluorescent signal proceeds exactly as in the case of incoherent excitation.[7] Here we summarize some of the results of that analysis and illustrate these results with some simple examples.

From a record of the fluorescent signal $I(t)$ it is possible to obtain the probability density $W_{\rm off}(\tau_{\rm off})$ for the durations $\tau_{\rm off}$ of interruptions in the fluorescence and the probability density $W_{\rm on}(\tau_{\rm on})$ for the durations $\tau_{\rm on}$ of intervals during which the fluorescence is on. The theory predicts distributions of the form[7]

$$W_{\rm off}(\tau_{\rm off})=R_-\exp(-R_-\tau_{\rm off})\ , \tag{30a}$$

$$W_{\rm on}(\tau_{\rm on})=R_+\exp(-R_+\tau_{\rm on})\ . \tag{30b}$$

It follows that the mean signal-off time is $\bar{\tau}_{\rm off}=1/R_-$ and the mean signal-on time is $\bar{\tau}_{\rm on}=1/R_+$. For example, if we tune to one component of the Autler-Townes doublet ($\Delta_2=\pm\frac{1}{2}\Omega_1$, $R_+=\Omega_2^2/2A_1$), then the mean time between interruptions in the fluorescence ($\bar{\tau}_{\rm on}=2A_1/\Omega_2^2$) becomes very large as the field strength tends to zero ($\Omega_2\to 0$), but the duration of the interruptions ($\bar{\tau}_{\rm off}=1/R_-$) tends to $1/A_2$ in this limit. This is quite reasonable. A weak field is slow to excite the weak transition, but once excited it decays in the natural lifetime $\tau_2=1/A_2$.

Another quantity accessible to experiment is the two-time intensity correlation function $\langle I(t)I(t+T)\rangle$. Here, as in earlier work, we assume that the photon-emission rate on the strong transition is so high that the fluorescent intensity at a nearby detector may be treated as constant [$I(T)=I_0$] when the fluorescence is on, and, of course, zero [$I(t)=0$] when it is switched off. With $I(t)$ defined in this way, the theory yields

$$\langle I(t)I(t+T)\rangle=m_I^2+\sigma_I^2\exp[-(R_++R_-)T]\ , \tag{31}$$

where $m_I=I_0R_+/(R_++R_-)$ is the mean intensity and

$$\sigma_I^2=\langle I^2\rangle-\langle I\rangle^2=I_0^2R_+R_-/(R_++R_-)^2$$

is the variance of intensity.[7] The most dramatic correlation effects occur when the fractional fluctuation of intensity $\sigma_I/m_I=(R_+/R_-)^{1/2}$ is large. This occurs when $R_+\gg A_2$, in which case $\sigma_I/m_I=2^{-1/2}$.

Observe that the intensity correlation function tends to $\langle I^2\rangle$ as $T\to 0$. This appears to violate the accepted result that the quantum-intensity correlation function for single-atom fluorescence tends to zero as $T\to 0$ (photon antibunching). The explanation for this apparent disparity is simply that the correlation function (31) assumes a coarse-grained time scale. In the quantum correlation function, the width of the antibunching dip at $T=0$ is of order A_1^{-1}. This is exceedingly narrow on the time scale of interest for intermittent fluorescence (say time units of order A_2^{-1}), and is not resolved in the present coarse-grained calculation. A more rigorous fully quantum-mechanical calculation shows that, for $T\gg A_1^{-1}$, the purely quantum-mechanical part of the intensity correlation (the part that gives rise to photon antibunching) is negligible, and only the classical part given by Eq. (31) remains. This justifies use of the classical intensity in the present calculation.

A more complete statistical description of the intermittent signal is provided by the probability $P_n^{\rm on}(t,T)$ that n weak transitions occur in the time interval $[t,t+T]$ with the fluorescence on at time $t+T$ and the probability $P_n^{\rm off}(t,T)$ that n weak transitions occur on $[t,t+T]$ with the fluorescence off at time $t+T$. The theory[7] indicates that these probabilities obey the equations

$$dP_n^{\rm on}/dT=R_-P_{n-1}^{\rm off}-R_+P_n^{\rm on}\ , \tag{32a}$$

$$dP_n^{\rm off}/dT=R_+P_{n-1}^{\rm on}-R_-P_n^{\rm off}\ , \tag{32b}$$

with initial conditions $P_{-1}^{\rm on}(t,T)=P_{-1}^{\rm off}(t,T)=0$ $P_0^{\rm on}(t,0)=\mathscr{P}_-(t)$, $P_0^{\rm off}(t,0)=\mathscr{P}_+(t)$, and $P_n^{\rm on,off}(t,0)=0$, for $n>0$.

The total number of weak transitions on $[t,t+T]$, i.e., the total number of times the fluorescence switches on or off, without regard for the final state of fluorescence, is

$$P_n(t,T)=P_n^{\rm on}(t,T)+P_n^{\rm off}(t,T)\ . \tag{33}$$

When the weak transition is strongly saturated ($R_+\gg A_2$) and the strong one is not ($\Omega_1\ll A_1$), the upward and downward rates, Eqs. (29), are nearly equal ($R_+=R_-=R$), and Eqs. (32) can be added to obtain an equation for P_n,

$$\frac{dP_n}{dT}=R(P_{n-1}-P_n)\ . \tag{34}$$

The solution of this equation is the Poisson distribution

$$P_n(T)=\frac{(RT)^ne^{-RT}}{n!}\ . \tag{35}$$

But for weak excitation of transition $0\leftrightarrow 2$ ($R_+\ll A_2$), the number of switches in time T does not have a Poisson distribution. This is because the upward transitions require a long period of time while the downward ones occur quickly ($\bar{\tau}_{\rm on}=1/R_+\gg\bar{\tau}_{\rm off}=1/A_2$). Hence the switches occur in pairs (switch off and switch on) separated by a relatively long period of time. This precludes a Poisson distribution.

IV. CONCLUSION

Starting from the density-matrix equation (1), we have obtained a statistical description of the random fluorescence from the strong transition. The essential feature of our treatment is the observation that the disparity of the time scales A_1^{-1} and A_2^{-1} allows a drastic reduction in the complexity of the problem to the set of equations (23). As we have pointed out, a sufficient condition for the validity of the analysis is that the rates $R_\pm$ be small compared to the rates of relaxation of atomic coherence ($\sigma_{01},\sigma_{02},\sigma_{12}$) and of populations of the $0\leftrightarrow 1$ transition. Relative to our previous work, a number of new features arise due to the coherent nature of the excitation, perhaps the most noticeable of which are the Autler-Townes splitting [Eq. (27)] and the increase in transverse relaxation rate of the weak transition [Eq. (20b)]. As in the case of incoherent illumination we have identified regimes in which the interruptions of the strong signal fluorescence due to transitions to and from level 2 dominate the character of this signal. Conversely, for excitation with a strong resonant field ($\Omega_1\gg A_1,\Delta_1=0=\Delta_2$), the interruptions become extremely rare, since the probability for excitation to level 2 is, in this case, approximately $\mathscr{P}_+\sim\Omega_2^2A_1^3/A_2\Omega_1^4\ll 1$ unless Ω_2/A_2 is itself of order A_1/A_2. Due to the Autler-Townes splitting, one is in effect driving the weak transition far from resonance.

Since our analysis relies upon a coarse-graining of time with respect to the lifetime of the strong transition (A_1^{-1}), we lose such features as photon antibunching of the light from the $0\leftrightarrow 1$ transition. However, from the point of view of the intermittent fluorescence, such features are not of principal importance, since their extent

in time is extremely small as compared to the time scale A_2^{-1} of the intermittent fluorescence for the situation we envision ($A_1/A_2 \gg 10^8$). It is the long time scale A_2^{-1} that is encoded in the strong fluorescence and which provides the remarkable character of the interruptions. Incidentally, one need not restrict attention only to optical transitions to produce intermittent fluorescence. Transitions induced by collisions would serve equally well as Cooper and Ballagh have shown.[15]

Of course, the light radiated by the atom and the sequence of photoelectric pulses that it would produce are not equivalent to the classical stochastic process that we have described as the resolution in time with which one views the system becomes increasingly fine. It is only the limit of times long compared to A_1^{-1} that the nonclassical character of the emission is lost. From this perspective it is of interest to inquire about the fashion in which one moves from a nonclassical to a classical process by time averaging. In a sense the choice of temporal resolution by the observer dictates the interpretation of and extent to which the processes on the strong transition are nonclassical. The situation is analogous to that encountered in resonance fluorescence from a two-level atom in which the choice by the observer of either frequency or time-resolved measurement leads to an interpretation in terms of a cascade through a ladder of dressed states or of projection into the "undressed" atomic ground state. In the present problem, the existence of intervals of darkness due to the excitation of the weak transition is a question largely independent of the choice of time scale. However, the question of the nonclassical nature of the fluorescence from the strong transition is certainly intimately related to the choice of time resolution.

Finally, we note that, since this paper was submitted, a number of related theoretical works have appeared,[16–20] the simplest and clearest approach being that of Cohen-Tannoudji and Dalibard.[16] Some of these works questioned the existence of dark periods in the fluorescent signal. The question was settled by the experiments of Bergquist *et al.*[21] and of Nagourney *et al.*[22] in which the intermittent fluorescence of single atomic ions was observed.

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